AD732519

THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS

A QUARTERLY TECHNICAL REPORT,

SUBMITTED TO

THE U.S. ARMY RESEARCH OFFICE

PERIOD

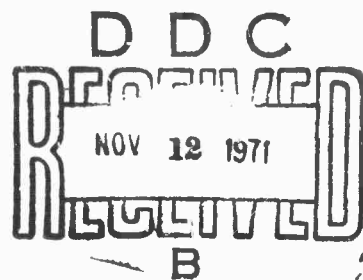June 22 to September 21, 1971

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RECEIVED
NOV 12 1971
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23

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)*	2a. REPORT SECURITY CLASSIFICATION
Department of Electrical Engineering University of Maryland College Park, Maryland 20740	Unclassified
	2b. GROUP

3. REPORT TITLE

The Study of the Interaction of Intense Picosecond Light Pulses with Material (Mode-Locked Lasers)

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*

A quarterly technical report, June 22 to Sept. 21, 1971

5. AUTHOR(S) *(First name, middle initial, last name)*

Chi H. Lee

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
October 15, 1971	18	24

8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)
DA-ARO-D-31-124-71-G55	
b. PROJECT NO. ARPA Order No. 675 Am. 9	
c. Program Code No. 9E20	9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)*
d.	

10. DISTRIBUTION STATEMENT

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11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	ARPA and U.S. Army Research Office

13. ABSTRACT

Recent advance in the development of mode-locked lasers is reviewed. In particular the prinicple of generation and the measurement of the picosecord pulses are discussed. Recent measurements on the spectral and temeral characteristics are included. This data can be used to explain some of the discrepancy found in previous measurements. The application of picosecond pulse for lifetime measurement in semi-conductor is also discussed.

DD FORM 1473
1 NOV 65

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14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Lasers						
Mode-locking						
Picosecond light pulses						

MODE - LOCKED LASERS

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Mode Locked Lasers

I. Introduction

Ultrashort pulse generated by mode-locking has caused much of the
interest in the area of quantum electronics recently. The unique charac-
teristics of such light pulse: the ultrshort time duration and the extreme
high peak power, have stimulated the investigation of many physical and
chemical processes at picosecond time scale. For example, it is now
possible to investigate the transient response of a physical system. The
electric field associated with the ultrashort optical pulse may approach the
atomic field of matter. One can thus expect to observe the higher order
nonlinear optical effects which up to now have not been reported.

The mode-locked lasers in general refered to a class of lasers which
utilize the principle of mode-locking to achieve ultrashort pulse generation.
The output usually consists of a train of such pulses with regular spacing.
In principle, all kind of lasers can be mode-locked by either active or passive
means. In the literature mode-locked solid state, gas, dye and even semi-
conductor lasers have been reported. All of them are based on the same
principle although individual type of laser may require specific design
consideration and possesses unique character. However, following historical
development, we shall put our emphasis on the discussion of the mode-locked
Nd:glass laser. Historically, it was due to the successful operation of a
self mode-locked Nd:glass laser developed by DeMaria and his coworker

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in 1966[1] that stimulated intensive experimental and theoretical studies in this field. Today, after half decade of research, we still lack complete understanding of the mode-locked Nd:glass laser.

II. Basic Operation Principle of the Mode-Locked Lasers.

The basic principle of the mode-locked laser operation is very simple. It is well known from Fourier theorem that any repetitive function with a definite period can be represented by a Fourier series. Let

$$E(t) = a_0 + \sum_{n=1}^{\infty} (a_n \cos \frac{2n\pi}{T} t) \tag{1}$$
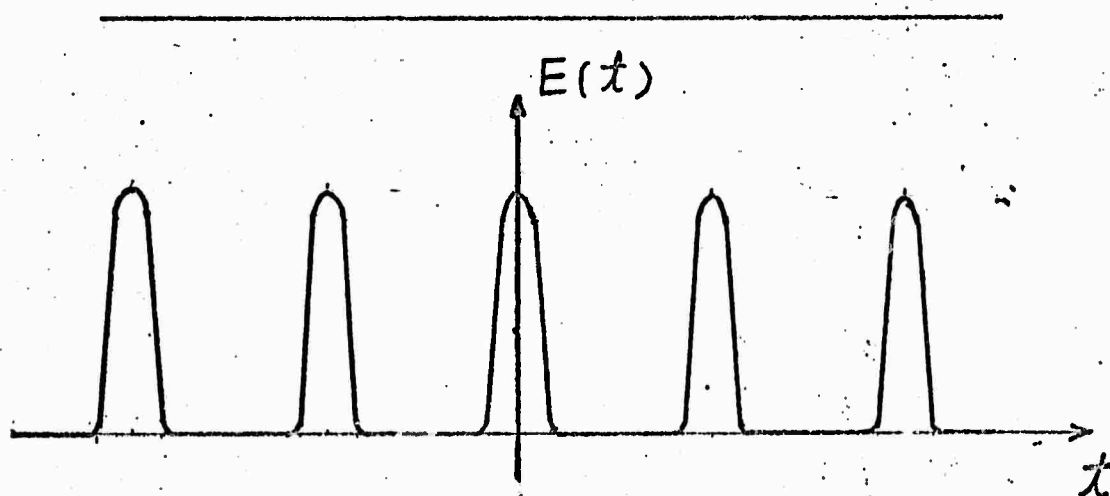
represent the function shown in Fig. 1.

$E(t)$

t

Fig. 1

where the expansion coefficients are given by

$$a_0 = \frac{1}{T} \int_{-T/2}^{T/2} E(t) dt \tag{2}$$

$$a_n = \frac{2}{T} \int_{-T/2}^{T/2} E(t) \cos \frac{2n\pi}{T} t \, dt \qquad n = 1, 2, 3, \ldots \tag{3}$$

Note that we have omitted the sine part of the series expansion because the

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function shown in Fig.1 is an even one. The value of the coefficients depends on the shape of the pulse. Let's consider two simple cases.

Case 1.

$$E(t) = \begin{cases} 0 & \text{when} & -2 < t < -1 \\ k & \text{when} & -1 < t < 1 \\ 0 & \text{when} & 1 < t < 2 \end{cases} \qquad (4)$$

$$E(t) = \frac{k}{2} + \frac{2k}{\pi} \left(\cos \frac{\pi}{2} t - \frac{1}{3} \cos \frac{3\pi}{2} t + \frac{1}{5} \cos \frac{5\pi}{2} t - + \cdots \right) \quad (5)$$

where the period $T = 4$

Case 2. $E(t)$ is a δ- function like pulse at $t = 0, T, \ldots nT, \ldots$
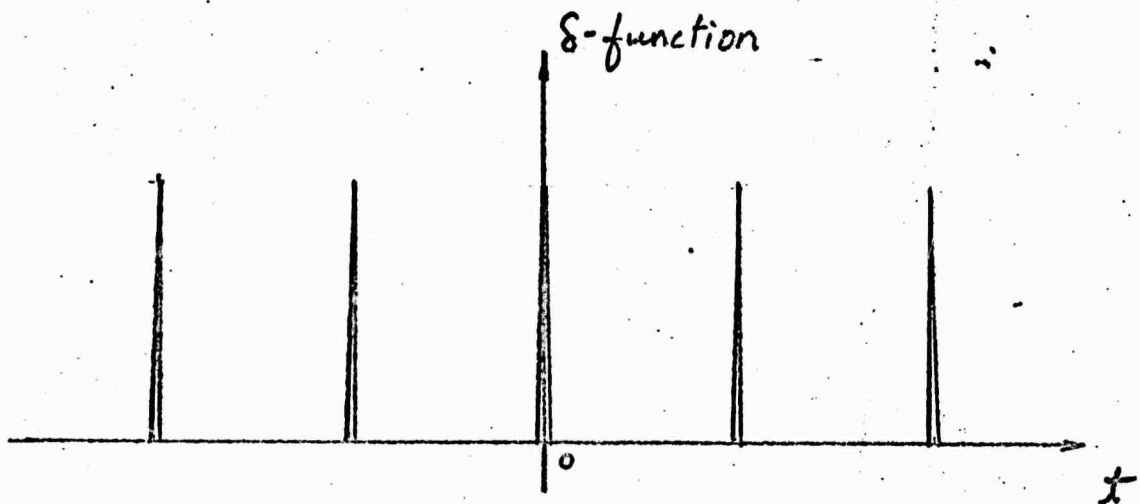
The pulse is shown in Fig. 2.

Fig. 2.

Simple calculation will yield

$$a_0 = \frac{1}{T} \int_{-T/2}^{T/2} \delta(t) \, dt = \frac{K}{T} \qquad (6)$$

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$$K = \int_{-T/2}^{T/2} \xi'(t)\,dt = \text{total area (or strength) of the } \delta\text{-function}$$

$$a_n = \frac{2}{T} \quad \text{for all } n = 1, 2, 3, \ldots \tag{7}$$

From these two examples one can draw the following qualitative statement. If we let $\cos\left(\frac{2n\pi}{T}t + \phi_n\right)$ denote the nth mode of the optical resonantor then the conditions that all these modes will interfere properly to give a ultrashort pulse train are the following

   (1) The modes must be in phase, i.e. $\phi_n = 0$ for all n

   (2) All modes are of equal amplitude.

We also make the observation that the more the number of modes are included the narrower the pulse width will be. For an δ-function like pulse, the number of mode required is infinite. In general the pulse width, $\Delta t \sim \frac{1}{M\Delta f}$ where M is the total number of modes and Δf is the mode spacing. For the Fabry Perot optical resonantor $\Delta f = \frac{c}{2L}$, where L is the effective optical length of the cavity. It is a simple matter to show that

$$\Delta f = \frac{1}{T} \quad \text{or} \quad T = \frac{2L}{c} \tag{8}$$

i.e., the period of the repetitive pulses is equal to the cavity round-trip transit time.

An ideal mode-locked laser is a good physical system to realize the mathematical expression shown in equation (1). In the case of a Nd:glass laser, the width of the laser gain curve can support the simultanueous oscillation of ten thousand cavity modes. What is needed is a mechanism to couple all the modes and equalize their phases. This phase-locking element may be added as follows. It is reasonable to assume that some

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mode at frequency ν_m near the peak of the laser gain profile will be oscillated first. If an amplitude modulator operating at a frequency f is inserted into the laser's feedback interferometer, the mode ν_m will be amplitude-modulated at a frequency f such that its time dependent electric field will be of the form

$$E(t) = E_m (1 + M \cos 2\pi ft) \cos 2\pi \nu_m t \tag{9}$$

where $\nu_m = m\Delta f$

or

$$E(t) = E_m \cos 2\pi \nu_m t + \frac{ME_m}{2} \cos 2\pi (\nu_m - f) t$$
$$+ \frac{ME_m}{2} \cos 2\pi (\nu_m + f)t. \tag{10}$$

One sees that new side-bands are generated. If the modulation frequency is equal to the mode spacing, i. e. $f = \Delta f$, then these new side bands will be able to grow. One sees that one starts with one mode and ends up with three modes all oscillating in-phase if the modulation frequency is correctly chosen. As the new oscillating modes pass through the modulator, they also become amplitude-modulated. Their side bands, in turn, couple the $\nu_m + 2\Delta f$, or ν_{m+2} and $\nu_m - 2\Delta f$ (ν_{m-2}) modes to the previous three modes. This process continues until all axial modes falling within the laser gain curve are coupled. In the actual operation of a mode-locked Nd:glass laser, the amplitude modulator is the saturable dye. A saturable absorber can be regarded as a two-level quantum system whose separation energy is equal to the photon energy of the incident light pulse. As the pulse propagates through the absorber, the leading edge of the pulse is heavily absorbed and atoms make transition from lower to upper state of the two-level system. As a result of this transition, the absorber tends to saturate. This process

represents the opening of the passive modulator. After the pulse has propagated through the system, the atoms make transitions to the lower state via relaxation, this represents the closing of the modulator. Some time later, all the atoms are in the lower state and the modulator is again closed. Thus the opening and closing of the absorber is atuomatically timed at the round-trip cavity transit time by the very circulating pulse itself. This process introduces an amplitude modulation to the initail pulse at the correct frequency, i.e. at $f = \Delta f = \frac{c}{2L}$ = mode spacing. The argument of the mode coupling process mentioned earlier will follow. Alternatively one can regard the nonlinear absorber as a pulse sharpener since it provides less attenuation for the higher-amplitude portions of the pulse and higher-attenuation for the lower-amplitude portions. The requirements for the saturable absorber are (i) that it has an absorption line at the laser wavelength (iii) that the dye's recovery time be shorter than the cavity round trip transit time. Such satrable absorbers are available for the Nd:glass laser. They are Eastman-9740 and 9860 reversible dye solutions. For the mode-locked ruby laser, the proper dye to use is the solution of cryptocyanine in acetone.

Analytical theory for the mode-locked laser has also been worked out by various authors. [2-6] In general one can describe the theory either in frequency domain or in time domain. For the frequency domain description, complete theory is available only for the situation involved three or four axial modes. Thus the theory is necessary to be qualitative. It has been shown by Tang and Statz [2] that in a laser operation there exists a definite phase relationships between modes. This is the so-called maximum-emission principle. According to this principle, the particular set of relative phases

that maximizes the total rate of stimulated emission is the one that becomes established in the laser. The basic physical mechanism that is responsible for the phase-locking effect is the nonlinear saturation in the laser medium or in the saturable absorber. For the Nd:glass laser, it has been shown both theoretically and experimentally that the strength of mode-locking is provided by the nonlinear saturation of the absorber. We can summarize the description of mode-locking in freuency domain as follows. The process begins with the oscillation of a few modes. The phases of these modes are locked by the maximum emission or minimum loss principle in either the active medium or the nonlinear absorber. The side bands are generated by the nonlinear absorption of the Q-switched dye. All the side bands thus generated will coincide with the cavity mode and are phase-locked. The process continues until all the modes under the laser gain curve are brought into oscillation and phase-locked. This ideal mode-locking will produce a train of pulses whose individual pulse width is equal to the inverse band width of the laser gain curve. In the case of the Nd:glass laser, the band width of the laser is about 100$\overset{\text{o}}{\text{A}}$ and it can support the simultaneous oscillation of 10^4 modes which, when all locked together, will produce pulse of 3×10^{-13} second. We like to point out once more that even though the frequency domain model of mode-locking is easy to understand, however the theoretical calculation is only possible for a very limited number of modes. The consequence of this is that we can never have a quantitative theory which can facilitate the direct comparison with the experiments. Such shortcoming is partially removed if one describes the process in the time domain.

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The time domain description of the mode-locking has been introduced by Basov (et al)[5] and Fleck[6]. Fleck's model is based on traveling-wave equations which are derived from the Maxwell's equations and solved in conjunction with boundary conditions imposed at the cavity mirrors. The coupled differential equations are then transformed into difference equations which are solved numerically by computer. The result describes the details of the simultanious Q-switched and mode-locked pulse evolution from noise. This model shows that the pulse evolution can be divided into three stages. In the initial stage, the intensity is low enough that both the amplification and absorption processes can be considered to be linear. The intensity pattern is that of amplified spontaneous emission and obviously represents Gausian random noise. Initially the intensity output is aperiodic, but as the radiation is amplified above noise background there are quasi-periodic similarities between the emission over different round trips. Since the laser exhibits net amplification, the radiation undergoes spectral narrowing. This effect is exhibited in the time domain as a smoothening and broadening of pulses existing in the round-trip patterns. In the second phase of the pulse evolution, the absorption is nonlinear but the amplification is linear. This phase ends when the absorbing transition is completely saturated. As the result of nonlinear absorption, two effects take place. First, there is a selective emphasis of certain of the pulses already present. Since the processes involved are frequency dependent, the selection may not always be on the basis of height alone. For ideal mode-locking, the number of pulses per round trip should be narrowed down to one. The second effect

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of nonlinear absorption is spectral broadening which tends to narrow the existing pulses in time. This effect is however partly counteracted by the tendency of the linearly amplifying region to narrow the spectrum. The final phase of the pulse evolution occurs when the intensity is sufficiently high for complete saturation of the absorber transition to take place and for the amplification to be nonlinear. It would be expected that the nonlinear amplification would further broaden the spectrum and narrow the temporal width of the pulse. However this does not happen. One would certainly expect nonlinear pulse narrowing in time to take place after the passage of radiation through a sufficiently long path in an amplifying medium. When the path is folded back and forth upon itself as it is within a laser cavity, however, the nonlinear pulse narrowing capability can be very much reduced if the pump cannot restore the inversion lost during each pass before the reflected pulse reenters the amplifying medium. This is the situation encountered with sold state lasers.

III. Measurement of Picosecond Light Pulses.

Conventional light pulse measurement is performed with the photo-diode or multiplier and the light pulse is converted into electronic pulse and displayed on an oscilloscope. The best photo diode - oscilloscope combination can resolve one tenth of a nanosecond. Wide band width measurement are available by a sampling scope, however such technique is only suitable to measure mode-locked pulse train of a cw laser. For a Q-switched and mode-locked output such as that derived froma Nd:glass or ruby laser, the sampling technique is not applicable for the obvious reason

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that there are not enough data points for sampling. It is clear that there is no direct measurement of the Q-switched and mode-locked picosecond pulses. Indirect measurement employing both linear or nonlinear optical effects are available in wide variety. All these indirect methods measure the autocorrelation functions in various order of the electric field. It should be pointed out that among them the linear technique similar to the Michelson's interferometer experiment as reported by Smith and Alley[7] is not suitable for the pulse width measurement since it measures the auto correlation function of the electric field. It is well known from Fourier analysis that the autocorrelation function of electric field can be obtained by Fourier transforming the power density spectrum of the pulse. Thus two measurements are identical and they give the lower limit of the pulse width not the pulse width itself.

Among all the techniques the ones that are more practical are multi-photon absorption-fluorescence and optical harmonic generation. Giordmaine et al[8] in 1967 reported the two photon absorption-fluorescence technique for the measurement of picosecond laser pulses. It measure the intensity correlation function of the pulse. Its simplicity is its major advantage over the harmonic generation techniques. In the two-photon absorption-fluorescence technique one splits the beam into two parts and recombines them in a fluorescence dye cell as shown in the triangular geometry in Fig. 3
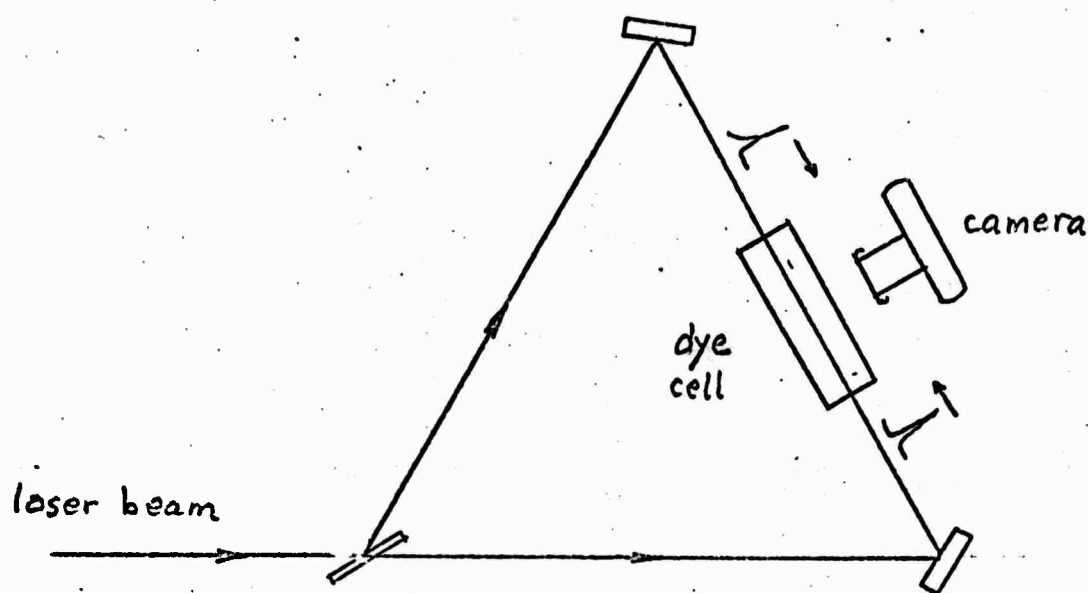
Fig. 3

The absorption of the dye used is peak at about twice the frequency of the light pulse. It is necessary to simultaneously absorb two light quanta in order to excite the dye from its ground state to higher electronic state. The absorption is thus proportional to the square of the intensity of the light beam. The dye after excited will decay back to its original level by accompanying fluorescence emission. The intensity of fluorescence emission is directly proportional to the absorption. Hence in the region where two beam overlapped, the fluorescence intensity will be strongest. By photographing the fluorescence track from the side of the cell, one will see a bright spot indicating the extent that the two beam overlapped. The spatial size of the bright spot is an indirect measurement of the time duration

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of the picosecond light pulses. Mathematically the photograph is a measure

of the expression call fluorescence yield.

$$f(\tau) = 1 + 2G^{(2)}(\tau)$$

where $G^{(2)}(\tau)$ is the second order autocorrelation function defined by:

$$G^{(2)}(\tau) = \frac{\int I(t) \, I(t+\tau) \, dt}{\int I^2(t) \, dt} \tag{12}$$

It has been pointed out by Weber[9] and Klauder et al[10] that extreme care

must be exercised in the interpretation of data since similar fluorescence

track will be obtained even if a non mode-locked laser beam is used. The

criteria to decide whether a laser is mode-locked and produces picosecond

pulse or not is based on the measurement of the contrast ratio of the bright

fluorescence spot to the background. For ideal mode-locked laser, the

contrast ratio is 3. The random phased, thermal noise type of laser out-

put will yield contrast ratio of 1.5. The difference is only a factor of two.

Therefore the usefulness of this technique is limited and for correct inter-

pretation of the data the accurate measurement of the contrast ratio must be

carried out. Using two photon fluorescence technique for the measurement

of Nd:glass lasers consistently give pulse measurement of 4 to 10 picosecond

with contrast ratio of 1.8 to 2.0. This imposes difficulty in understanding

the mode-locking process and the characteristics of the Nd:glass laser

since two very important points can not be resolved: (1) The pulse width

measured is wider than what the inverse of the laser band width will give,

(2) The contrast ratio is very much less than the theoretically predicted value

of 3 for the mode-locked pulses. Such discrepency has stimulated intensive

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studies both theoretically and experimentally. A review in this problem is provided by Duguay, Hansen and Shapiro. [11]

Another less ambiguous but more elaborated method is the harmonic generation technique. Armstrong[12] was the first one to report on the pico-second pulse measurement by using reflected second harmonic generation in GaAs crystal. He obtained 8 picosecond as the duration of the pulse. Recently, Eckardt and Lee[13] have used the optical third harmonic generation method to measure the pulse. The improved resolution of the technique makes it possible to reveal the subpicosecond structure of the pulse. The result agree quite well with the improved technique based on two-photon fluorescence method.

IV. Characteristics of the Mode-Locked Nd:glass Lasers:

As we pointed out before that there was no direct measurement on the mode-locked pulses. All temporal characteristics of the pulses had to be inferred indirectly from measurements. In this section we just summarize the up-to-date finding about the pulses from some typical mode-locked Nd:glass lasers. We sort out this particular type of laser because of its complexity. Other types of cw mode locked laser are relatively simple to understand because in the mode-locking process no complication has been induced. For a mode-locked Nd:glass laser it has been found that the pulse width is always one order of magnitude wider than the inverse of the laser band width would allow. To account for this discrepency, Treacy[14] has reported the observation of frequency chirping of the pulse. Picard and Schweitzer[15] also proposed a domain locking model to account both for the

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two photon fluorescence trace and the contrast ratio measured. It is

generally agreeable now that the picosecond pulse actually consists

of subpicosecond structure whose width is equal to the inverse band

width of the laser. However the detail shape of the pulse evelop of

the picosecond light pulse is still unknown. More recently, Korobkin

et al[16], Duguay et al[11] and Eckardt, Lee and Bradford[17] have

reported the observation of self-phase modulation of the pulse due to

intensity dependent index of refraction of the laser medium. Eckardt

et al used a ring laser cavity to study in detail how the spectral and

temporal evolution of the pulse train are. They found that the mode-

locked pulses evolve from a very narrow spectrum width of $4 \overset{o}{A}$ to the

full extent of $80 \overset{o}{A}$. The spectral broadening observed there is quite

different in mechanism than that due to the mode-coupling. The spectral

broadening is due to modulation of the phase by the index change. If

the pulse intensity is strong enough, this same effect will also lead

to the self-focusing of the beam and thus cause spatial energy

redistribution. The data seems to indicate that the side-band generation

are mainly due to the self-phase modulation while the coupling of the

neighboring axial mode is due to nonlinear absorption. The intensity

dependent index of refraction can also lead to chirping of the pulse

since at the peak intensity of the pulse the index will be larger. This

portion of the pulse will accordingly travel at a slower velocity than

the other part of the pulse and cause the wavelength at the leading edge

to stretch and the trailing edge to compress. It is clear that a complete

theory of mode-locking should include the intensity dependent index change
in the laser medium.

We like to conclude this section by pointing out that all the nonlinear
techniques for pulse measurement can only give information about the auto-
correlation function of various order. It is well known that one can not
construct the original pulse by just knowing the correlation function. How-
ever, in principle if the correlation functions of various order can be measured
simultaneously, then the original pulse can be constructed. This process will
certainly be tedious if not impossible. The best measurement of the pulse
therefore rests upon the real time display of the pulse. Such device is
possible in the future since the state of the art technique of using modified
image converter camera can already provide 6 picosecond resolution time.[18]

V. Applications of Picosecond Laser Pulses.

There are numerous applications of the picosecond light pulses. Due
to their ultrshort duration, such pulses are ideal for probing the atomic or
molecular event which has the relaxation time of a few picoseconds. It also
offers extreme accuracy in optical ranging, high speed photography. It is
particularly suitable as the light source for nonlinear optical study since,
on one hand it can provide extreme high peak power which is needed for the
observation and, on the other hand, the short duration of the pulse causes
the material damage threshold moving up. Many nonlinear optical effects
which are otherwise nonobservable are now possible to detect with the use
of picosecond light pulses. Such examples can be found in the case of third
harmonic generation in reflection from semiconductor surface and the

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transient stimulated Raman effect. [19] Complete theory has been worked
out for the latter case. The regime of interest for the transient effect is one in
which the laser and/or Stokes pulses have a duration shorter than the
dephasing time of the excited state of the molecules. The transient stimulated
Raman effect differs from the steady state one in the sense that the gain of
the transient effect depends only on the total Raman-scattering cross section,
while the steady-state gain is inversely proportional to the line width. It has
been shown experimentally that in methanol two lines (at $2837cm^{-1}$ and
$2942cm^{-1}$) can be stimulated while in the steady-state case only the stronger
and narrower $2837cm^{-1}$ line can be stimulated. Many other liquids and gases
also show transient stimulated Raman effect. The efficiency of the conversion
to Stokes frequency is as high as 40%. This effect thus provides a convenient
way to generate picosecond pulses at various wavelengths. It has also been
shown that stimulated Raman scattering from picosecond pulses can give
rise to Stokes pulses of considerable shorter duration then that of the initiating
pulses. In the earlier experiment a single pass Raman oscillator was employed.
Such pulse narrowing was not observed due to the group velocity mismatch
between the Stokes and the pump pulses causing the reduction of gain. Recently,
Colles[20] showed that the Stokes pulse narrowing did take place if one used
a multiple reflection cavity for the short pulse Raman oscillator and the cavity
length was adjusted to compensate the mismatch so that a simple Stokes
pulse bouncing back and forth in the Raman oscillator was amplified by
successive pump pulses. With this technique, the width reduction of one
order of magnitude was observed. The peak power of the Stokes pulse can

also exceed that of the pump pulse, a situation in analogy to the backward Stimulated Raman Scattering.

Picosecond pulses are also good for studying the transient response of the photo-excited charge carriers in semiconductor. Recently, Jayaraman and Lee[21], in their study of two-photon conductivity effect in GaAs crystal have used picosecond mode-locked pulse train from an output of a Nd:glass laser as the excitation source. They have observed the response of the conductivity of the semiconductor in accordance with the modulation of the pulse train. This gives a direct observation of the lifetime of the charge carrier to be shorter than the separation of the adjacent pulses, which is about 5 nanosecond in their case. In the study of photoconductivity effect, the high peak power of the light pulse is also responsible to the observation of the higher order effects, such as stimulated hole absorption and three photon absorption in these crystals.

The high intensity and short duration of mode-locked pulses make them, ideal for a number of different type of lifetime measurements. For picosecond time resolution the technique first demonstrated by Shelton and Armstrong[22] may be used. The experimental method used to measure such short decay time is as follows. Absorption of a very intense light pulse of ultrashort duration by a dilute sample of dye molecules will prepare all the molecules in the light path in an excited electronic state. Such pulse may be called as the "preparing" pulse. The subsequent decay of the population of this state is then probed by a "probing" pulse which can be delayed continuously to arrive before, during or after the intense "preparing" pulse.

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The transmission of the sample for the probe pulse is inversly proportional at each instance to the concentration of the ground state dye molecules. As the sample is prepared in the excited state, the transmission will rise abruptly. As the ground state repopulated through decay from the excited state, transmission will decrease again. A measure of the width of the transmission curve verses delay time will give the decay time of the dye. Using this technique, Shelton and Armstrong have reported the lifetime of the Q-switched dye at 25 picoseconds or less.

Picosecond light pulses also prove to be valuable for generating plasma and as scientific tools in controlled thermonuclear research. Basov and Krokhin[23] calculated that laser power in excess of 10^9 watts was needed to heat a laser generated lithium deuteride plasma up to a temperature at which thermonuclear neutron emission may be observed. However 10^9 watts power for a nanosecond pulse proves to be too much for any optical component to withstand. Optical damage to the components resulting from such large optical intensity made it difficult to carry out such experiments af that time. It was subsequently noticed that the power damage threshold increased with decreased pulse duration. The possibility of reaching thermonuclear neutron emission temperatures with high intensity, ultrashort pulse is clear. The use of an Nd:glass single picosecond pulse generator in conjunction with five amplifying stages has result in the generation of 20 joule of energy in ten picosecond. Experimental results of the use of these high-energy, single ultrashort pulses in generating thermonuclear neutron emission from laser heated lithium deuteride surfaces[24] have been reported subsequently by several groups.

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